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STATE OF WASHINGTON

DEPARTMENT OF ECOLOGY

7601 W. Clearwater, Suite 102 • Kennewick, Washington 99336 • (509) 546-2990

July 7, 1994

Mr. J.D. Bauer, Program Manager
Office of Environmental Assurance, Permits, and Policy
U.S. Department of Energy
P.O. Box 550
Richland, WA 99352

Mr. R.E. Lerch, Deputy Manager
Restoration and Remediation
Westinghouse Hanford Company
P.O. Box 1970
Richland, WA 99352

Dear Messrs. Bauer and Lerch:

Re: Tank 241-CX-72 at the Strontium Semiworks

Following receipt of your October 25, 1993, letter to Dru Butler, Hanford Facility Dangerous Waste Part A Permit Application Form 3. Revision 2. for the 241-CX Tank System (WA7890008967), questions were raised regarding the safety of tank CX-72 and the advisability of deferring sampling of the sludge in this tank.

The Washington State Department of Ecology (Ecology) has concluded its review of the conditions in Tank CX-72, and now agrees to the deferral of sampling until work on the 200-SO-1 operable unit begins. We also accept the Part A Permit Application as complete. For your information, I am including a copy of the "Safety Assessment of Hanford Strontium Semiworks Tank 241-CX-72," prepared for Ecology by our contractor, Black & Veatch.

To ensure that conditions in Tank CX-72 do not deteriorate, I am requesting that the following measures be taken until the 200-SO-1 workplan deems otherwise:

- Maintain the building over Tank CX-72 in its present condition
- Prohibit the use of this building for any purposes other than the current one
- Preserve access to the drywell in tank CX-72
- Conduct monthly inspections to verify compliance with the above conditions

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With these conditions met, Ecology is satisfied that the waste in Tank CX-72 can continue to be safely stored until waste removal occurs in conjunction with the 200-SO-1 site remediation. Under milestone M-20, Ecology would have expected a closure plan for the 241-CX Tank System by May 1996. Before integrating this closure plan into the operable unit workplan, Ecology and USDOE must agree on a new milestone for submittal of the workplan. Please submit a proposed milestone for completion of the 200-SO-1 workplan to Ecology and the U.S. Environmental Protection Agency no later than August 8, 1994.

If you have any questions about this matter please call Ms. Nancy Uziemblo of my staff at 736-3014.

Sincerely,

David L. Lundstrom
Nuclear Waste Program

DL:NU:mf

cc: Doug Sherwood, EPA
Administrative Record (200-SO-1)

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plutonium mass and multiplication factor which are likely to be higher than is actually the case.

We appreciated the opportunity to provide these services for the Department of Ecology. John Kirkland assisted with this work by doing the majority of the criticality calculations. We also had help, and at times, advice, from Dr. M. John Robinson, former faculty member at Kansas State University (KSU)and presently Partner-in-Charge of Nuclear-related projects for Black & Veatch.

Should you require clarification or additional information, or should you have any questions or comments, please contact us.

Very truly yours,

BLACK & VEATCH

Ernest L. Seth

Ernest L. Seth

cc: L. F. Drbal
R. M. Prewitt
J. C. Kirkland
E. L. Seth/File
B. Bailey

BLACK & VEATCH

Washington Department of Ecology
Hanford Waste Management

B&V Project 40478.020
B&V File 15.0200
May 5, 1994

9414200.2760

Washington Department of Ecology
Nuclear Waste Program
P. O. Box 47600
Olympia, WA 98504-7600

Subject: Safety Assessment of Strontium Semiworks Tank 241-CX-72

Attention: Mr. Mike Gordon

Gentlemen:

Attached is the final report on the safety assessment of Hanford Strontium Semiworks Tank 241-CX-72. As stated in the report, assuming the nuclide masses you supplied and worst case conditions of a homogeneous water-heavy metal mixture in the 11-foot section of the tank now containing waste, calculated values of k_∞ range from about 0.30 to about 0.45. Please note that the density is a function of the fluoride-oxide mixture percentages, with a 30% oxide mixture yielding a density of nearly 27 grams per cubic centimeter. Since pure uranium and plutonium metals have densities of less than 20 g/cc, this does not seem a credible value.

Applying the information about the metal-salt and -hydroxide content of the "average tank" resulted in a maximum Pu-239 mass of about 1475 grams with a density of 2.26 g/cc, equating to about 1.19% oxides with k_∞ of 0.37. Bear in mind that k_∞ would be reduced by the addition of mass necessary to account for two-thirds of the waste material in the form of such compounds as $NaNO_3$. The addition of these constituents was not considered in the calculation of the multiplication factor because the quantities are unknown.

Granting the assumptions made, the final result is that the effective multiplication factor, k_{eff}, is likely to be less than 0.1 in the dry condition, as seems to be the case at present, and could be no higher than the maximum value of k_∞ noted above. As you are undoubtedly aware, the methods used are conservative and result in values for

SAFETY ASSESSMENT

HANFORD STRONTIUM SEMIWORKS TANK 241-CX-72

for

WASHINGTON DEPARTMENT OF ECOLOGY

April 1994

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SUMMARY

An analysis was performed to estimate the potential for nuclear criticality under worst case conditions in Strontium Semiworks Tank 241-CX-72. A mixture of transuranic nuclides equal to the average mixture previously calculated to be in other single shell tanks at the Hanford site was used as the basis for calculation. Worst case conditions were assumed to be those in which the heavy metals present were homogeneously mixed with water in the 11-foot section of the tank now containing essentially dry waste. Calculations of the infinite multiplication factor were carried out using the full range of combinations of both the fluoride and oxide forms. Results indicate that the infinite multiplication factor lies between values of 0.30 and 0.45. Given the assumptions and the geometry of the problem, the effective multiplication factor, and therefore the potential for an unintentional criticality event in the tank, is sufficiently low as to indicate no further concern for such an event. Additional safety considerations beyond those indicated in the report, such as the possibility of leakage of the contained waste material, lie outside the scope of this report.

1.0 Introduction

The assignment for this project was to review the adequacy of existing documentation on the waste characterization for the Strontium Semiworks Tank 241-CX-72 (Tank CX-72), review and comment on the safety/criticality of the tank and contained waste, and to evaluate the quality of the U.S. Department of Energy(DOE)/Westinghouse Hanford Company (WHC) plutonium inventory estimates for the material in Tank CX-72.

Documents were supplied (References 1 & 2) which supplied the basis for the previous DOE/WHC estimates of tank plutonium inventory. Reference 1 presents historical information intended to support the results obtained and documented in Reference 2. In addition, total radionuclide inventory for single-shell tanks at the Hanford site was supplied by the Washington Department of Ecology (Ecology) as an excerpt from "Tank Waste Technical Option Report," WHC-EP-0616 (Reference 3). On March 29, 1992, a meeting was held at the Federal Building in Richland, Washington, among representatives of Ecology, DOE, WHC and Black & Veatch at which some additional information was presented.

Based on the supplied information and the above referenced documents, reviews of the aforementioned documentation, tank safety and criticality, and DOE/WHC plutonium inventory estimates were conducted.

Questions which were asked by the Washington Department of Ecology included:

1. How likely is a criticality event in the tank?
2. Are the DOE-supplied documents and the methods they present sufficient to answer question 1?
3. Are the consequences of a criticality event significant in this tank?
4. What alternatives can be pursued to reduce the uncertainties?
5. What information can be gained from the proposed "active neutron interrogation?"
   a. Can moisture content be inferred?
   b. Try to anticipate a cut-off value below which there is some assurance that there will be no criticality problem.

It was believed that the first question, above, was of greatest concern to Ecology, and that a result indicating that such an event is very unlikely would reduce or eliminate the necessity for concern related to the others. In the light of the foregoing, a study was performed to attempt to determine the likelihood of a criticality event in Tank CX-72. As is shown in Figures 1 and 2, if certain assumptions are accepted, a criticality in the tank is an extremely unlikely event.

The methods, assumptions and results are detailed in the following sections, as are more detailed conclusions which were drawn from these results. These results should provide answers to the above questions which will allow Ecology to reach an informed decision regarding future actions relating to Tank CX-72.

2.0 Document Review

The work represented by References 1 & 2 appears to be sound. It is recognized that most of the information needed to gain a clear-cut, verifiable, understanding of the physical processes occurring in the waste is unknown. Records necessary for a complete understanding of the chemistry and physics of the waste material do not exist. In the absence of data, assumptions must be made in order to carry out even a simplified investigation. References 1 and 2 attempted to 'fill in the blanks' in the needed data by both historical and radiological means.

In the "Discussion" section of Reference 1, it is pointed out that the accuracy of some of the historical information "...is highly suspect." This is attributed to several reasons, among them the elapsed time and the nature of the record keeping at the time. Also noted were the following:

1. Whether or not decontamination flush waste was stored in Tank CX-72 is still a question.

2. Stated inventories of radionuclides are rough estimates and cannot be verified.

3. Although the compositions of the chemicals used in the separation processes and decontamination were documented, there is no documentation of the amounts used.

4. The quantities, concentrations and current chemical compositions of the materials in the tank are unknown.

Because of the many unknowns and the lack of information about the waste, radiological measurements were needed before a decision could be made regarding the need to remove the waste, sample the tank, apply other means of investigation or store the waste in place. Many of these measurements were made by WHC, resulting in Reference 2.

Reference 2 is a report on an attempt to characterize the waste in the tank by radiological analysis. The methods used in the study appear to be sound, though the scarcity of information relating to Ecology's concerns makes it understandable that questions would remain. From the standpoint of the questions asked by Ecology, the only points at which results presented in Reference 2 may be questioned are linked by the alpha-n cross sections. These are the assumption that the chemical composition of all the transuranics is the fluoride form, and the lack of information regarding criticality considerations. The 'all fluorides' assumption, while probably not far removed from possibility, is not conservative and is difficult to justify on the basis of the information reported in Reference 2. It would appear that the criticality question was not considered at the time the study was done and therefore must have been outside the scope of the

study. Given no other information about the basis for the nonconservative assumption, one would expect that the assumption itself, as well as the resulting low value for plutonium content, would certainly be questioned in subsequent criticality investigations based upon the work of Reference 2.

The results of the measurements reported in Reference 2 provide some indication of the radiological processes occurring in the tank and place some constraints on existing conditions in the waste. For instance, the waste is stable, obviously not critical and, as indicated by the low number of thermal neutrons found, seems to be a dry solid.

The waste material appears to be confined to the bottom 10 to 11 feet of the tank, capped by a grout layer which extends to the top of the tank. Radiological measurements strongly indicate a waste - grout interface at a height of approximately 11 feet above bottom. This, assuming a 40-inch tank diameter, allows a determination of the maximum volume of waste of

$$\text{Vol} = \pi r^2 h = \pi(20 \text{ in} \times 2.54 \text{ cm/in})^2(11.0 \text{ ft} \times 12 \text{ in/ft} \times 2.54 \text{ cm/in})$$

$$\text{Vol} = \pi(50.80 \text{ cm})^2(335.28 \text{ cm}) = 2.718 \times 10^6 \text{ cm}^3.$$

3.0 Safety and Criticality

Radiological safety concerns can not be limited to the criticality issue. It should be noted that, according to Reference 2, various chemicals were used as solvents and decontamination agents. These chemicals could attack the steel wall of the tank, over the years, resulting in the potential for loss of integrity of the primary containment barrier. The question of the possibility of hazardous material, such as chromium, being contained in the waste in tank CX-72 is beyond the scope of this study.

This section of the report, for the convenience of the user, will first present definitions of the notation, followed by the assumptions and data used in the following calculations. Next, based upon the supplied data, a range of representative radionuclide compositions of the waste material will be developed, followed by an analysis of criticality over that range of compositions.

3.1 Notation and Definitions

N atomic number density (atoms · cm^{-3}). The number of atoms of a particular isotope per unit volume. Also used as the total number of atoms.

GAW gram atomic weight. The mass, in grams, of one mole of atoms of an element or isotope.

b barn. A unit of cross section measurement. 1 b = 10^{-24} cm^2.

σ_i microscopic cross section (b) where i represents either *a* for absorption or *f* for fission. The probability that a neutron-nuclear reaction of this type will occur.

Σ_i macroscopic cross section (cm^{-1}) where i represents either *a* for absorption or *f* for fission. The product of an atom density and a microscopic cross section. The total number of events of the given type which occur within the unit volume.

I_i resonance integral (b) where i represents either *a* for absorption or *f* for fission. Essentially a microscopic cross section average over all resonance regions.

ν fission multiplicity (unitless). The number of neutrons released from fission, either neutron induced or spontaneous fission.

k_∞ infinite multiplication factor. The ratio of the number of neutrons produced to the number of neutrons absorbed in an infinite system.

t.f. subscript to denote thermal fission.

s.f. subscript to denote spontaneous fission.

(α,n) a nuclear reaction in which an alpha particle is absorbed by a nucleus and as a result emits a neutron. Also used as a subscript to denote an (α,n) reaction.

$t_{1/2}$ radioactive half life. The time required for one-half of a radioactive nuclide to decay to its daughter product.

λ radioactive decay constant. The quotient of the natural logarithm of 2 and the radioactive half-life.

N_A Avogadro's Number. 6.022×10^{23} atoms per gram-atom

3.2 Assumptions

The following assumptions, along with a brief explanation of each, are used here and should be noted.

1. Two groups of cross sections are considered. 2200 $m \cdot s^{-1}$ cross section data are used to approximate reactions in the thermal range. Resonance integrals are used to approximate cross sections for all energies above thermal. In effect, the resonance integral is treated as an average cross section for resonance levels.

2. When calculating resonance fissions, the value of the thermal induced neutron multiplicity will be used. This is done both for convenience and because insufficient data on ν for resonance energies was found.

3. While arguments can be made both for and against the use of average tank values of radionuclide inventory in this study, the decision was made to use the single-shell tank radionuclide inventory from WHC-EP-0616 as the basis for further criticality evaluation. The composition of the waste assumed to be contained in Tank CX-72 is, therefore, that provided by the Washington Department of Ecology, from Tank Waste Technical Options Report WHC-EP-0616 (Reference 3). Only the heavy transuranic nuclides and the constituents of oxides, fluorides and water (O_2, F_2, H_2) will be considered in this analysis. Exclusion of other materials is conservative in the determination of Σ_a. For the purpose of this study, a conservative scenario was chosen in which all fissile and fissionable material is contained in a homogeneous water solution. This results in a conservative upper limit on the infinite multiplication factor.

4. Tank CX-72 is assumed to have a radius of 20 inches and a waste layer height of 11 feet, resulting in a volume of 2,718,000 cm^3. This is consistent with the DOE/WHC documentation which was supplied and with Department of Ecology assumptions.

5. A volumetric neutron production rate of 7 $n \cdot s^{-1} \cdot cm^{-3}$ is assumed, which is consistent with the WHC radiological study of Tank CX-72. Taking tank volume into account, this results in a total neutron production rate of 1.9026×10^7 $n \cdot s^{-1}$.

3.3 Data

The data presented in Tables 3.3-1 and 3.3-2 were used in the calculations shown in following sections of this report. Microscopic cross sections were taken from Table 19, pg. 2-22, of Reference 6. Thermal fission multiplicity (ν) and spontaneous fission yield are from Table 11-1, pg. 339, of Ref. 5. (α,n) yields for oxides and fluorides are from Table 11-3, pg. 345, of Ref. 5.

Table 3.3-1. Neutron Yields [Ref. 5]

	$\text{yield}_{s.f.}$ $(n \cdot s^{-1} \cdot g^{-1})$	$\text{oxide yield}_{(\alpha,n)}$ $(n \cdot s^{-1} \cdot g^{-1})$	$\text{fluoride yield}_{(\alpha,n)}$ $(n \cdot s^{-1} \cdot g^{-1})$
^{233}U	8.60×10^{-4}	4.8	7.0×10^{2}
^{234}U	5.02×10^{-3}	3.0	5.8×10^{2}
^{235}U	2.99×10^{-4}	7.1×10^{-4}	8.0×10^{-2}
^{238}U	1.36×10^{-2}	8.3×10^{-5}	2.8×10^{-2}
^{237}Np	1.14×10^{-4}	3.4×10^{-1}	--
^{238}Pu	2.59×10^{3}	1.34×10^{4}	2.2×10^{6}
^{239}Pu	2.18×10^{-2}	3.81×10^{2}	5.6×10^{3}
^{240}Pu	1.02×10^{3}	1.41×10^{2}	2.1×10^{4}
^{241}Pu	5.0×10^{-2}	1.3	1.7×10^{2}
^{242}Pu	1.72×10^{3}	2.0	2.7×10^{2}
^{241}Am	1.18	2.69×10^{3}	--
^{242}Cm	2.10×10^{7}	3.76×10^{6}	--
^{244}Cm	1.08×10^{7}	7.73×10^{4}	--

Table 3.3-2. Cross Section Data [Ref. 5, 6]

	σ_a (b)	σ_f (b)	I_a (b)	I_f (b)	$\nu_{t.f.}$
^{233}U	581	527	900	746	2.4
^{234}U	97.5	0.65	665	-----	2.4
^{235}U	694	582	370	240	2.41
^{238}U	2.71	-----	19.9	-----	-----
^{237}Np	170	0.019	756	-----	2.7
^{238}Pu	403	16.6	175	25	2.9
^{239}Pu	1026	746	430	130	2.88
^{240}Pu	295	0.1	2000	-----	2.8
^{241}Pu	1400	1025	678	537	2.8
^{242}Pu	30	0.2	1280	0.6	2.81
^{241}Am	630	3.1	2150	-----	3.09
^{242}Cm	20	5	-----	-----	3.44
^{244}Cm	15	-----	660	12.5	3.46
^{1}H	0.33	-----	0.166	-----	-----
^{16}O	0.0002	-----	0.00027	-----	-----
^{19}F	0.009	-----	0.0176	-----	-----

3.4 Estimation of Tank CX-72 Contents

The contents of Tank CX-72 are a mixture of several materials. For the purpose of determining tank contents, the assumption will be made that the waste in Tank CX-72 consists only of a homogeneous mixture of water and the following thirteen radionuclides: the uranium isotopes 233, 234, 235, and 238, the neptunium isotope 237, the plutonium isotopes 238, 239, 240, 241, and 242, the americium isotope 241, and the curium isotopes 242 and 243.

Activities of these isotopes, as well as the other materials not considered, were provided by the Washington Department of Ecology. These activities are representative of the contents of similar single-shell tanks located at the Hanford Reservation. The activities of the thirteen radionuclides are included in Table 3.4-1, along with the half life and decay constants of the individual isotopes. The decay constant is calculated by

$$\lambda_i = \frac{ln(2)}{t_{\frac{1}{2},i}}, \qquad (3.4\text{-}1)$$

(Ref. 4, pg 14) where $t_{\frac{1}{2},i}$ is the half-life and λ_i the decay constant of the ith radionuclide. In calculating decay constants, one year is assumed to be equal to

$$1\ y = (356.25\ d \cdot y^{-1}) \times (24\ h \cdot d^{-1}) \times (3600\ s \cdot h^{-1}) = 3.15576 \times 10^{6}\ s.$$

The activity of an isotope can be used to determine the mass of the material. The activity of any radioactive material is defined by

$$A = N\lambda, \qquad (3.4\text{-}2)$$

(Ref. 4, pg 13) where A is the activity in disintegrations per second (assuming that λ is in units of s^{-1}) and N is the number of atoms of the radioactive material. Mass is related to the number of atoms of a material by the expression

$$m = \frac{N \times GAW}{N_A}. \qquad (3.4\text{-}3)$$

Table 3.4-1. Representative Composition of Tank

Nuclide	$t_{1/2}$, Half-life	λ (s^{-1})	Activity (Ci)
^{233}U	1.59×10^{5} years	1.3814×10^{-13}	5.7×10^{-3}
^{234}U	2.45×10^{5} years	8.9651×10^{-14}	1.6×10^{-1}
^{235}U	7.04×10^{8} years	3.1200×10^{-17}	2.0×10^{1}
^{238}U	4.47×10^{9} years	4.9138×10^{-18}	4.7×10^{2}
^{237}Np	2.14×10^{6} years	1.0264×10^{-14}	6.0×10^{1}
^{238}Pu	87.74 years	2.5034×10^{-10}	1.2×10^{3}
^{239}Pu	2.41×10^{4} years	9.1139×10^{-13}	2.1×10^{4}
^{240}Pu	6.56×10^{3} years	3.3482×10^{-12}	5.1×10^{3}
^{241}Pu	14.35 years	1.5306×10^{-9}	5.2×10^{4}
^{242}Pu	3.76×10^{5} years	5.8416×10^{-14}	2.0×10^{-4}
^{241}Am	433.6 years	5.0656×10^{-11}	4.1×10^{4}
^{242}Cm	163 days	4.9218×10^{-8}	5.4×10^{1}
^{244}Cm	18.1 years	1.2135×10^{-9}	1.6×10^{2}

Since the activities in Table 3.4-1 are given in Curies, Eq. 3.4-3 can be modified to yield

$$m = \frac{A\times3.7\times10^{10}\times GAW}{N_A\lambda}, \qquad (3.4\text{-}4)$$

where A is in Curies, and one Curie is defined as 3.7×10^{10} disintegrations per second.

With a known mass of each radionuclide, the mass fraction of each may be determined. Due to the assumption that these materials are the only tank constituents, the mass fraction of a nuclide is the ratio of the isotopic mass to the total mass in the tank. Table 3.4-2 summarizes the masses required to yield the activities given in Table 3.4-1, and the mass fraction of each isotope. As Table 3.4-2 indicates, ^{238}U dominates the tank contents (99.3%), with significant amounts of ^{235}U (0.66%) and ^{239}Pu (0.02%) present.

The contents of Tank CX-72 are contributing neutrons by spontaneous fissions and (α,n) reactions with fluorine or oxygen. Since the neutron yields for these two processes are given in $n \cdot s^{-1} \cdot g^{-1}$, the neutron production rate $(n \cdot s^{-1} \cdot)$ in the tank is

$$\dot{R} = \sum_{i=1}^{13} m_i (Y_{s.f.} + Y_{\alpha,n}), \qquad (3.4\text{-}5)$$

where the summation index i represents the thirteen radionuclides contained in the tank and Y indicates the neutron yield of each of the two processes. Since the total mass is the sum of the individual constituents, and the total neutron production rate is known (see Section 3.2, #5), Eq. 3.4-5 simplifies to

$$\dot{R} = \sum_{i=1}^{13} f_i m_{tot} (Y_{s.f.} + Y_{\alpha,n}) \quad \rightarrow \quad m_{tot} = \frac{1.9026 \times 10^7}{\sum_{i=1}^{13} f_i (Y_{s.f.} + Y_{\alpha,n})}, \qquad (3.4\text{-}6)$$

where f_i is the mass fraction of the ith nuclide.

The spontaneous fission yields and the (α,n) yields for oxide fuels and fluoride fuels are given in Section 3.3. The mass fractions for each constituent were calculated as shown above and are included in Table 3.4-2.

Tables 3.4-3 and 3.4-4 summarize the results of solutions of Equation 3.4-6 assuming these materials are 100% oxides and 100% fluorides, respectively. The two tables indicate the value of each term in the summation of Eq. 3.4-6, and the mass of each individual isotope. The total mass was solved using Eq. 3.4-6, and the individual masses obtained by multiplying the total mass by the appropriate mass fraction.

In the case of an all oxides mixture, the total waste mass is 2.362×10^8 g, with 1.552×10^6 g of ^{235}U, 2.346×10^8 g of ^{238}U, and 5.675×10^4 g of ^{239}Pu. The total mass results in a material density of 86.914 $g \cdot cm^{-3}$.

In the case of an all fluorides mixture, the total waste mass is 3.384×10^6 g, with 2.224×10^4 g of ^{235}U, 3.361×10^6 g of ^{238}U, and 8.131×10^2 g of ^{239}Pu. The total mass results in a material density of 1.245 $g \cdot cm^{-3}$.

Since the radionuclide inventory from Reference 3 indicates a density of 1.67 $kg \cdot L^{-1}$ (1.67 $g \cdot cm^{-3}$), this would indicate that the majority of the tank contents are fissile materials in fluoride form.

Table 3.4-2. Mass Fractions of Tank Contents

Nuclide	mass (g)	fraction of total mass
^{233}U	5.907006×10^{-1}	4.19411×10^{-10}
^{234}U	2.565910×10^{1}	1.82185×10^{-8}
^{235}U	9.255715×10^{6}	6.57177×10^{-3}
^{238}U	1.398691×10^{9}	9.93103×10^{-1}
^{237}Np	8.512416×10^{4}	6.04401×10^{-5}
^{238}Pu	7.009633×10^{1}	4.97700×10^{-8}
^{239}Pu	3.383558×10^{5}	2.40240×10^{-4}
^{240}Pu	2.246078×10^{4}	1.59477×10^{-5}
^{241}Pu	5.030507×10^{2}	3.57177×10^{-7}
^{242}Pu	5.090640×10^{-2}	3.61447×10^{-11}
^{241}Am	1.198477×10^{4}	8.50946×10^{-6}
^{242}Cm	1.631350×10^{-2}	1.15829×10^{-11}
^{244}Cm	1.976641×10^{0}	1.40346×10^{-9}
Total	1.408405×10^{9}	1.00

Table 3.4-3. Tank Inventory Assuming All Oxides

Nuclide	$f\times(Y_{sf}+Y_{\alpha n})$	mass in tank (g)
^{233}U	2.013534×10^{-9}	9.90788×10^{-2}
^{234}U	5.474696×10^{-8}	4.30382×10^{0}
^{235}U	6.630916×10^{-6}	1.55247×10^{6}
^{238}U	1.358860×10^{-2}	2.34604×10^{8}
^{237}Np	2.055652×10^{-5}	1.42784×10^{4}
^{238}Pu	7.958000×10^{-4}	1.17573×10^{1}
^{239}Pu	9.158400×10^{-3}	5.67527×10^{4}
^{240}Pu	1.851530×10^{-2}	3.76738×10^{3}
^{241}Pu	4.821889×10^{-7}	8.43771×10^{1}
^{242}Pu	6.224117×10^{-6}	8.53858×10^{-3}
^{241}Am	2.290050×10^{-2}	2.01022×10^{3}
^{242}Cm	2.867926×10^{-4}	2.73627×10^{-3}
^{244}Cm	1.526590×10^{-2}	3.31544×10^{-1}
Total	8.053900×10^{-2}	2.36233×10^{8}

Table 3.4-4. Tank Inventory Assuming All Fluorides

Nuclide	$f\times(Y_{sf}+Y_{an})$	mass in tank (g)
^{233}U	2.935881×10^{-7}	1.41947×10^{-3}
^{234}U	1.056682×10^{-5}	6.16595×10^{-2}
^{235}U	5.277000×10^{-4}	2.22418×10^{0}
^{238}U	4.131300×10^{-2}	3.36110×10^{6}
^{237}Np	6.890172×10^{-9}	2.04556×10^{2}
^{238}Pu	1.096229×10^{-1}	1.68444×10^{-1}
^{239}Pu	1.345349×10^{0}	8.13079×10^{2}
^{240}Pu	3.511684×10^{-1}	5.39741×10^{1}
^{241}Pu	6.073795×10^{-5}	1.20885×10^{0}
^{242}Pu	7.192795×10^{-6}	1.22330×10^{-4}
^{241}Am	3.758145×10^{0}	2.87998×10^{1}
^{242}Cm	2.432409×10^{-4}	3.92017×10^{-5}
^{244}Cm	1.515740×10^{-2}	4.74994×10^{-3}
Total	5.621598×10^{0}	3.38445×10^{6}

3.5 Criticality in Tank CX-72

Criticality of any nuclear system is defined by the effective multiplication factor, k_{eff}. This factor is the ratio of the number of neutrons produced to the number of neutrons absorbed, and corrected for the number of neutrons which leak out of the "core". Leakage is highly dependent upon the geometry of the system under investigation. When neutron leakage is neglected, the effective multiplication factor is referred to as the infinite multiplication factor, k_∞. k_∞ is the measure of k_{eff} in an infinite core, and is the ratio of the number of neutrons produced to the number of neutrons absorbed.

In general, the criticality of a reactor is determined by the numerical value of k_{eff}. When k_{eff} equals 1.0 the reactor is critical. A value of less than 1.0 is subcritical, and greater than 1.0 is supercritical. The same analogy is used for the tank by assuming that the tank and its contents form a homogeneous reactor core.

While k_{eff} determines the critical condition of a reactor, k_∞ can indicate whether the reactor can ever go critical. Since k_∞ assumes a reactor with no leakage, if k_∞ is less than or nearly equal to 1, the composition and geometry of the core materials make it impossible for the reactor to go critical. As an example of typical values for k_∞, the Kansas State University TRIGA Mark II reactor is a 20% enriched, 250 kW pool type reactor, and the value of k_∞ is approximately 1.38.

Therefore, k_∞ can be used as a basis for estimating criticality potential in Tank CX-72. Since leakage is ignored, k_∞ is a function of material only, and is determined by

$$k_\infty = \sum_i \frac{\nu \Sigma_f}{\Sigma_a}, \qquad (3.5\text{-}1)$$

(Ref. 4, pg 211, modified for multiple nuclides) where Σ_f and Σ_a are of the macroscopic cross sections of each of the constituents in the tank, ν is the multiplicity defined earlier, and the summation is over i materials. The macroscopic cross section is a function of the number density of an isotope, and is calculated by

$$\Sigma_i = N_i \sigma_i, \qquad (3.5\text{-}2)$$

(Ref. 4 pg 20). Equation 3.5-2 is also valid for resonance integrals when the assumption is made that the resonance integral is an effective microscopic cross section for neutrons across the resonance energy spectrum. Thus Eq. 3.5-2 can be modified to

$$\Sigma_i = N_i \times (\sigma_i + l_i) \qquad (3.5\text{-}3)$$

The number density, N, is the number of atoms of a material per unit volume. Thus the number density of nuclide *i* can be calculated by

$$N_i = \frac{m_i \times N_A}{GAW \times V}, \qquad (3.5\text{-}4)$$

As is seen in Eq. 3.5-4, the number density is a function of the mass of an isotope, which was determined in Section 3.4.

The worst-case scenario in Tank CX-72 would be that in which the entire volume containing the waste is filled with water. In other words, all of the constituent materials would be dissolved in water occupying a volume of 2.718×10^6 cm^3.

Equations 3.5-1 through 3.5-4 were used to estimate the value of k_∞ in the tank with all transuranic materials as oxides and with all as fluorides in the worst-case scenario. Tables 3.5-1 and 3.5-2 summarize the respective results of these calculations.

It should be noted that in the oxide form, all heavy materials are assumed to be in the form XO_2. Thus for every atom of a transuranic material, there are two atoms of oxygen. In the fluoride form, uranium is in the form UF_6 and plutonium is in the form PuF_4. The neptunium, americium, and curium isotopes are assumed to be in the same chemical form as plutonium. As is seen from the absorption cross section for fluorine in Table 3.5-2, the contribution is small. Hydrogen and oxygen are present in both cases due to the presence of water. Assuming the density of water is 1.0 g·cm^{-3}, there is a number of molecules of water equal to Avogadro's number per cubic centimeter of volume. With a gram molecular weight of 18, this leads to a number density of 6.691×10^{22} atoms·cm^{-3} of hydrogen, and 3.346×10^{22} atoms·cm^{-3} of oxygen in the tank due to the presence of water.

Table 3.5-1 indicates that a tank containing only transuranic oxides yields a k_∞ of 0.4565, well below what would be required for criticality. The tank containing an all-fluorides mixture yields a k_∞ of 0.3017, which is lower still.

Equations 3.5-1 through 3.5-4 can be used to calculate k_∞ for mixed oxide-fluoride materials in the tank, since the total masses of each form are already known. The mass of each of the materials is directly related to the percentage of that particular form in the tank. For example, in Section 3.4, it was determined that the mass of waste consisting of 100% oxides would be 2.362×10^8 g, while that consisting of 100% fluorides is 3.384×10^6 g. If an analysis were desired based on the assumption that the tank contains 50% of each chemical form, the total mass in the tank would then be found to

be m = $(0.5\times2.362\times10^{8}) + (0.5\times3.384\times10^{6})$ g = 1.198×10^{8} g. With the mass of each nuclide and its mass fraction known, Eqs. 3.5-1 through 3.5-4 may be used to determine k_∞.

Table 3.5-3 summarizes the value of k_∞ as a function of the oxide percentage present in the tank. The table lists oxide percentage ranging from 100% to 0% in 5% increments, density of tank contents (total mass divided by volume) and k_∞. Graphically, Figure 1 shows how k_∞ varies with the density of the tank contents. k_∞ is reasonably constant down to about 15 $g\cdot cm^{-3}$, then sharply declines. The shape of the graph is consistent with the behavior of k_∞.

Table 3.5-4 summarizes the value of k_∞ as a function of oxide percentage in a more narrow band. These results are for oxide percentages from 20% to 0% in 1% increments. This table and its accompanying Figure 2 are more representative of the probable fluoride-oxide mixture in the tank. Considering that uranium (which is the most abundant element in the tank) has a metal density of about 19 $g\cdot cm^{-3}$, it is unlikely that the density would be much higher than that value. A density of 19 $g\cdot cm^{-3}$ corresponds to a k_∞ of approximately 0.4475. A density of 10.0 $g\cdot cm^{-3}$ (which is the approximate density of UO_2) would yield a k_∞ of approximately 0.4375. Finally, a density of 1.67 $g\cdot cm^{-3}$ (which is the average density of the material found in tanks considered in Reference 3) would lead to a k_∞ of approximately 0.3375, while a density equal to dry sodium nitrate ($NaNO_3$ - the constituent which comprises more than two-thirds by weight of the average tank waste), 2.26 $g\cdot cm^{-3}$, would yield a k_∞ of about 0.37.

Table 3.5-1. Infinite Multiplication Factor Assuming All Oxides

Nuclide	N (# · cm^{-3})	N · ν · (σ_f+I$_f$) (cm^{-1})	N · (σ_a+I$_a$) (cm^{-1})
^{233}U	8.283734×10^{13}	2.530847×10^{-7}	1.226821×10^{-7}
^{234}U	3.584790×10^{15}	5.592273×10^{-9}	2.733403×10^{-6}
^{235}U	1.288259×10^{21}	2.552067×10^{0}	1.370708×10^{0}
^{238}U	1.925143×10^{23}	-----	4.352747×10^{0}
^{237}Np	1.175998×10^{19}	6.032867×10^{-7}	1.088974×10^{-2}
^{238}Pu	9.647953×10^{15}	1.163929×10^{-6}	5.576517×10^{-6}
^{239}Pu	4.639899×10^{19}	1.170591×10^{-1}	6.755693×10^{-2}
^{240}Pu	3.068752×10^{18}	8.592506×10^{-7}	7.042786×10^{-3}
^{241}Pu	6.847834×10^{16}	2.994969×10^{-4}	1.422980×10^{-4}
^{242}Pu	6.904407×10^{12}	1.552111×10^{-11}	9.044773×10^{-9}
^{241}Am	1.631444×10^{18}	1.562760×10^{-5}	4.535415×10^{-3}
^{242}Cm	2.212584×10^{12}	3.805644×10^{-11}	4.425168×10^{-11}
^{244}Cm	2.661481×10^{14}	1.756578×10^{-7}	1.796500×10^{-7}
^{1}H	6.691111×10^{22}	-----	3.318791×10^{-2}
^{16}O	4.211865×10^{23}	-----	1.979577×10^{-4}
Total		2.669445×10^{0}	5.847016×10^{0}

Table 3.5-2. Infinite Multiplication Factor Assuming All Fluorides

Nuclide	N (# · cm^{-3})	N · ν · (σ_f + I$_f$) (cm^{-1})	N · (σ_a + I$_a$) (cm^{-1})
^{233}U	9.063335×10^{11}	2.769030×10^{-9}	1.342280×10^{-9}
^{234}U	3.925654×10^{13}	6.124021×10^{-11}	2.993312×10^{-8}
^{235}U	1.412004×10^{19}	2.797208×10^{-2}	1.502372×10^{-2}
^{238}U	2.115583×10^{21}	----	4.783333×10^{-2}
^{237}Np	1.447969×10^{17}	7.428079×10^{-9}	1.340819×10^{-4}
^{238}Pu	1.188549×10^{14}	1.433866×10^{-8}	6.869815×10^{-8}
^{239}Pu	5.718913×10^{17}	1.442813×10^{-3}	8.326737×10^{-4}
^{240}Pu	3.784335×10^{16}	1.059614×10^{-8}	8.685050×10^{-5}
^{241}Pu	8.448984×10^{14}	3.695248×10^{-6}	1.755699×10^{-6}
^{242}Pu	8.523092×10^{10}	1.915991×10^{-13}	1.116525×10^{-10}
^{241}Am	2.012897×10^{16}	1.928154×10^{-7}	5.595854×10^{-5}
^{242}Cm	2.731298×10^{10}	4.697833×10^{-13}	5.462596×10^{-13}
^{244}Cm	3.288739×10^{12}	2.170568×10^{-9}	2.219899×10^{-9}
^{1}H	6.691111×10^{22}	----	3.318791×10^{-2}
^{16}O	3.345556×10^{22}	----	1.572411×10^{-5}
^{19}F	1.278132×10^{22}	----	3.399831×10^{-4}
Total		2.941880×10^{-2}	9.751210×10^{-2}

Table 3.5-3. Effect of Varying Oxide Percentage on k_∞ (Wide Range)

Percentage of Oxides	Density of waste ($g \cdot cm^{-3}$)	k_∞
100 (all oxides)	86.9144100	0.4565482
95	82.6309400	0.4564123
90	78.3474900	0.4562616
85	74.0640300	0.4560936
80	69.7805700	0.4559052
75	65.4971100	0.4556920
70	61.2136500	0.4554492
65	56.9301800	0.4551698
60	52.6467400	0.4548453
55	48.3632700	0.4544636
50	44.0798100	0.4540080
45	39.7963400	0.4534547
40	35.5128800	0.4527689
35	31.2294200	0.4518960
30	26.9459600	0.4507478
25	22.6625000	0.4491697
20	18.3790400	0.4468639
15	14.0955800	0.4431772
10	9.8121180	0.4363385
5	5.5286560	0.4192867
0 (all fluorides)	1.2451950	0.3016939

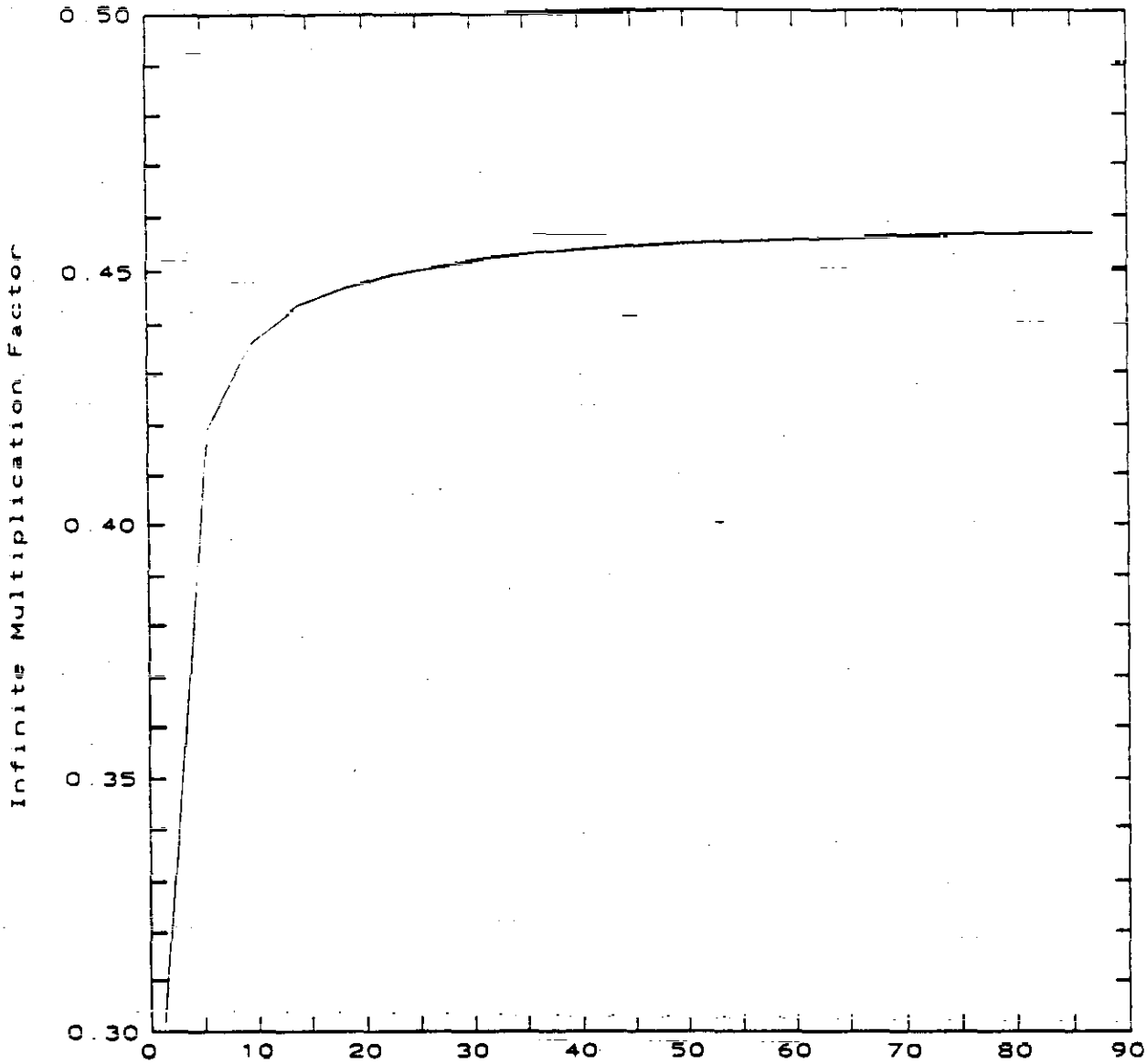

Figure 1. Multiplication vs. Density, Wide Range

Table 3.5-4. Effect of Varying Oxide Percentage on k_∞ (Narrow Range)

Percentage of Oxides	Density of waste ($g \cdot cm^{-3}$)	k_∞
20	18.3790400	0.4468640
19	17.5223500	0.4462691
18	16.6656600	0.4456140
17	15.8089600	0.4448885
16	14.9522700	0.4440812
15	14.0955800	0.4431773
14	13.2388900	0.4421581
13	12.3822000	0.4410002
12	11.5255000	0.4396732
11	10.6688100	0.4381372
10	9.8121190	0.4363385
9	8.9554270	0.4342037
8	8.0987350	0.4316282
7	7.2420420	0.4284606
6	6.3853500	0.4244696
5	5.5286580	0.4192868
4	4.6719660	0.4122840
3	3.8152730	0.4022989
2	2.9585810	0.3869105
1	2.1018890	0.3601065
0 (all fluorides)	1.2451970	0.3016941

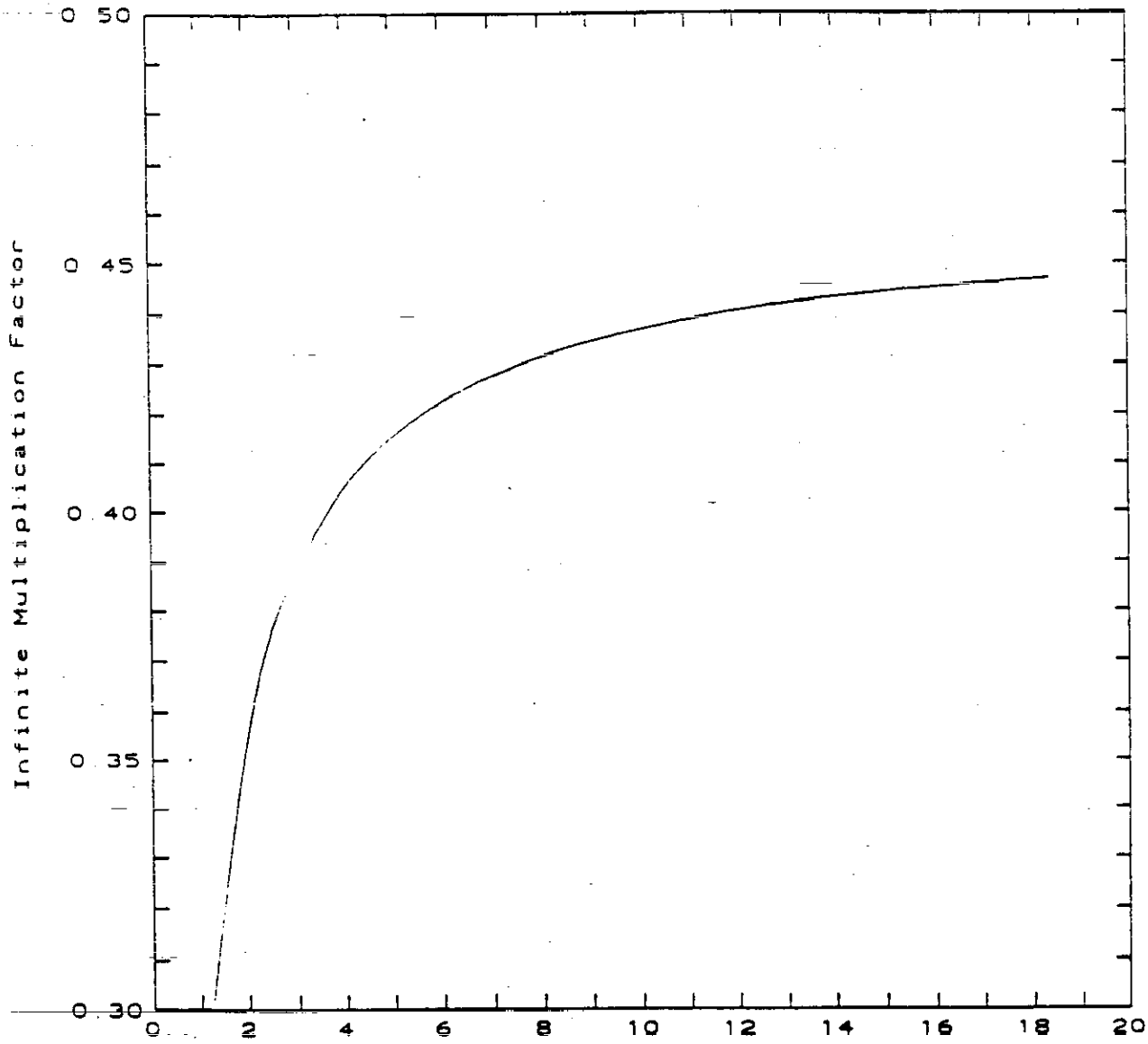

Figure 2. Multiplication vs. Density, Narrow Range

4.0 Results and Conclusions

4.1 How likely is a criticality event?

In the above attempt to gain some insight into the criticality situation in Tank CX-72, it was first assumed that the mass fractions of the waste in the tank are similar or close to the average of the single-shell tanks considered in Reference 3. While changes in mass fraction would result in changes to the indicated values of k_∞, the relationship is not linear. It is not possible to analyze the result of such changes without first assuming some changed mass fraction values, which may or may not more closely reflect reality.

The second assumption was that the mixture of heavy metals formed a homogeneous mixture with water. This is so unlikely as to be nearly impossible. Complete mixing would require not only the presence of a sufficient amount of water in the tank, but also a means of breaking up the solidified mass and mechanically mixing it. Simply adding water to the tank, which is already filled with solid material, could not add enough water to the sludge to drive the waste into the range of k_∞ shown in Figures 1 and 2. Assuming that there is little thermalizing material in the tank, whether water or something else, such as hydrocarbons, thermalized neutrons would be relatively rare. This was exactly the result found in the radiological study, Reference 2. For criticality to occur, then, would require a fast-fission process. Granting the geometry, the materials present and their mass fractions, the chance of a fast-fission criticality is nearly non-existent.

Next, consider the density of the waste. Pure plutonium and uranium metals have densities in the range of 19 grams per cubic centimeter. This density results in a conservatively calculated value of k_∞ of < 0.45. Oxides and fluorides have densities around 10 g/cc, yielding k_∞ < 0.44. The average density of waste (Reference 3) is about 1.67, producing k_∞ < 0.36. Assuming that the density of the waste is somewhat greater than the average does not greatly increase the value of k_∞. For instance, the average single-shell tank waste found in Reference 3 is at least 20% water. Dewatered, this waste would consist largely of $NaNO_3$, which has a density of about 2.26 g/cm^3. (Though the physical state of CX-72 waste is unknown, the low number of thermal neutrons found in Reference 2 indicates that it contains much less water than does the average waste tank.) Assuming that the density of waste in tank CX-72 is comparable to those of dry sodium salts or metal hydroxides, or about 2.26 grams/cm^3, rather than that of the average single-shell tank, results in a mixture of 1.19% heavy metal oxides (98.81% fluorides), with a total ^{239}Pu content of about 1,475 grams and k_∞ of 0.37. In this case, it appears safe to assume that the amount of oxides in the tank amounts to less than 5%, and is probably less than 1%, given the presence of other material which certainly is in the tank, though in unknown quantities, and has been neglected in this analysis. This neglected material will drive k_∞ lower because of the addition to total absorption.

Also of a conservative nature in the calculated values was the complete neglect of other materials in the tank, such as the salts and hydroxides mentioned above. While the species and amounts of such materials cannot be determined, Reference 3 does indicate

the presence of small amounts of samarium and technetium, both neutron absorbers which would drive k_∞ down by a small amount. Elements other than poisons in the system would still have a negative effect on k_∞ because of their contributions to total neutron absorption and their lack of neutron contribution through either fission or the alpha-neutron reaction. Simply stated, the addition of materials other than those shown in Section 3 would increase the denominator of the fraction shown in Equation 3.5-1 without affecting the numerator, thus resulting in a reduction in k_∞.

The use of k_∞ as a measure of criticality ignores neutron leakage. Leakage is important because neutrons which leak from the system under analysis cannot cause further reactions, and thus cease to be of interest to the analysis. Leakage is extremely important to criticality studies in its negative effect. A rough, non-analytical, estimate of the amount of leakage which might occur may be gained by the ratio of surface area to volume. In this case, Tank CX-72 is a relatively thin, long cylinder. Leakage here should be relatively high - likely 25% and possibly much higher. This means that k_{eff} is likely to be lower than k_∞ by at least 25%.

Because of the mixture involved, the geometry and the low values of k_∞, even assuming the most favorable conditions for initiation of such a reaction, there appears to be no plausible combination of events which might result in an unintentional criticality in Tank CX-72.

4.2 Are the DOE/WHC-supplied documents and methods sufficient to answer the above question?

As noted in Section 2, the documents supplied are of good quality. The studies appear to have been meticulously carried out. The results were not sufficient in themselves to eliminate the possibility of unintentional criticality because these studies did not include criticality analyses, and the reports lacked information which would be necessary to a determination of the probability of criticality. The radiological assessment reported in Reference 2 contains what appears to be at least one nonconservative assumption to arrive at a low value for the mass of plutonium-239 in the tank, which may have been used without question in subsequent criticality studies.

4.3 Are the consequences of a criticality event significant?

Unintentional criticality must always be considered a significant occurrence; one to be avoided. The consequences of a criticality event would vary, depending upon several factors. These factors include, but are not limited to,

(1) the amount of energy released,
(2) the rate of energy release,
(3) the condition of the tank wall, caisson and concrete footer.

Due to the lack of information regarding the tank contents, it must be understood that any assumptions made or conclusions drawn about the results of a criticality event in this situation are completely speculative. The range of possible responses of such a system cannot be estimated from the data supplied. Some general statements about such an event can be made, however.

Any criticality event will be accompanied by a rapid increase in both neutron and gamma fluxes. The magnitude of the radiation increase at the exposed tank top cannot be estimated, but could be expected to be measurable and would be primarily from gamma radiation. Most of the energy release is likely to be in the form of heat. The increase in temperature could be expected to cause increases in pressure due to expansion of volatile materials. Again, estimates of the magnitudes of the general and local temperature and pressure increases would be speculative.

The final result of the energy release would be one of three possibilities. The first is that the material would not allow an energy release rate sufficient to damage its container or cause a significant change of state of the waste material. The second is that the energy release could cause a rupture of the tank wall and a subsequent rearrangement of the geometry, resulting in a geometry which is not critical, and which cools slowly over a period of time. The third possibility is that the material could remain critical following a geometric rearrangement. The third possibility is the least likely of the three.

4.4 What alternatives can be pursued to reduce the uncertainties?

There are alternatives which may help to reduce uncertainties. One of these is the proposal to lower a neutron source into the dry well and measure the response of the system. This subject will receive more detailed attention in Section 4.5, below.

The suggestion was made to place monitors on the tank top or in the dry well to measure and record the gamma radiation level and to alarm if the flux exceeds a preset level. This would certainly provide a measure of safety to personnel in the vicinity and would provide the earliest possible alert following a hypothetical criticality event. It cannot be expected to provide an early warning of an impending criticality. An additional consideration, given the speculation of a monitored unintentional criticality, is what could be done to avoid or mitigate the criticality, even if such early warning were received. The answer to this question appears to be that personnel would be evacuated and the appropriate measures taken to best safeguard the health and safety of the public, in both the short and long terms.

Other alternatives are, of course, sampling and emptying the tank, either of which appear to be expensive.

4.5 What information can be gained from the proposed "active neutron interrogation?"

The proposed neutron interrogation, in which a neutron source of known strength would be lowered into the dry well, potentially could lead to accurate estimation of the effective neutron multiplication value of the system as it exists. This would include the effects of neutron absorbing materials in the waste, leakage from the system and neutrons from all sources, including those from neutron-alpha reactions and spontaneous fission from all sources in the waste. Thermal neutron flux measurements could be made during the course of the experiment, allowing inferences to be made about the amount of thermalizing material in the waste. This will not indicate the presence of water because, while it may indicate hydrogen content in the waste, other materials which may be present, can also reduce neutron energies toward the thermal region. It should be recalled, however, that previous measurements (Reference 2) have indicated that very few thermal neutrons were present.

This procedure will not provide an indication of how much of any particular transuranics may be present, nor relative measurements, such as weight percents. It will not provide information about the extent of the poisons which may be present. The presence or absence of water may be inferred, but not confirmed.

Neutrons emitted as a result of the various neutron-alpha reactions will be indistinguishable from those which result from fission, whether spontaneous or not. An indication of alpha-n activity may be gained, though not quantized, by the fact that the number of neutrons produced by alpha-n reactions will remain constant throughout.

The establishment of a 'cut-off' value of either k_∞ or k_{eff} is a difficult question. Any attempt to set such a value would amount to conjecture. The foregoing analysis would seem to render this a moot point, as the likelihood of criticality is so remote.

4.6 Conclusions

Granting that the assumption regarding relative abundances of the various nuclides is reasonably accurate, the calculated value of k_∞ precludes a criticality event in Tank CX-72. The calculated multiplication factor range assumes that worst-case conditions occur, which does not appear to be a credible possibility. k_{eff} can be expected to be much lower than that calculated here for several reasons, including the following:

a) No credible mechanistic scenario, other than intentional intervention, exists which would homogenize the tank contents with water.

b) There is no source of sufficient water to unintentionally flood Tank CX-72, assuming reasonable precautions are taken and/or procedures followed.

c) Both neutron leakage and the presence of neutron absorbers in the system were neglected in the calculation.

The present value of k_{eff} in the tank can be expected to be 0.1 or less. With such a low multiplication factor, the neutron source required to produce meaningful results from an active neutron interrogation would be large. Such an active source would complicate the procedure safeguards and increase the cost for little, if any, gain. While many uncertainties exist, the foregoing considerations cause doubt about the cost-benefits of such a procedure.

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